

# WORLD INTELLECTUAL PROPERTY ORGANIZATION International Bureau



#### INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 6	:		a	1) International Publication Number	: WO 96/37281			
B01D 53/86, A61L 9/20		A1		(43) International Publication Date: 28 November 1996 (28.				
(21) International Application Number:	PCT/US	96/063	05	(81) Designated States: AU, CN, JP,	KR, SG, European patent (AT, R, GB, GR, IE, IT, LU, MC,			
(22) International Filing Date:	3 May 1996 (	03,05.9	6)		R, OB, OR, IE, 11, DO, MC,			

(30) Priority Data:

Ţ

2,150,320

26 May 1995 (26.05.95)

CA Published

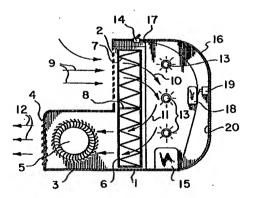
(71) Applicant: MINNESOTA MINING AND MANUFACTUR-

ING COMPANY [US/US]; 3M Center, P.O. Box 33427, Saint Paul, MN 55133-3427 (US).

(72) Inventors: DISANAYAKA, Bimsara; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). DIVIGALPITIYA, Ranjith; P.O. Box 33427, Saint Paul, MN 55133-3427 (US). LIV-INGSTONE, David, E.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US).

(74) Agents: GWIN, Doreen, S., L. et al.; Minnesota Mining and Manufacturing Company, Office of Intellectual Property Counsel, P.O. Box 33427, Saint Paul, MN 55133-3427 (US). ed With international search report.

(54) Title: BENCH TOP UV-ACTIVATED ODOR FILTRATION DEVICE



#### (57) Abstract

\*

The invention relates to an apparatus for purifying air by means of an activated photocatalyst such as titanium dioxide, the apparatus comprises a housing (1) having inlet (2) and outlet (4) for circulating air, a circulating means (5) such as a fan, is mounted in the housing to circulate air, a planar filter (7) disposed across the path of circulating air, the filter including a photocatalyst fixed to a fibrous porous support, and a light source (13) mounted in the housing to activate the photocatalyst; the invention provides a compact apparatus which can readily fit on a table or laboratory bench, and can quickly reduce air-borne contaminants such as odoss.

# FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AM	Armenia	GB	United Kingdom	MW	Malawi
AT	Austria	GE	Georgia	MX	Mexico
ΑU	Australia	GN	Guinea	NE	Niger
BB	Barbados	GR	Greece	NL	Netherlands
BE	Belgium	HU	Hungary	NO	Norway
BF	Burkina Faso	TE.	Ireland	NZ	New Zealand
BG	Bulgaria	IT	Italy	PL	Poland
BJ	Beain	JP	Japan	PT	Portugal
BR	Brazil	KE	Kenya	RO	Romania
BY	Belarus	KG	Kyrgystan	RU	Russian Federation
CA	Canada	KP	Democratic People's Republic	SD	Sudan
CF	Central African Republic		of Korea	SE	Sweden
CG	Congo	KR	Republic of Korea	SG	Singapore
CH	Switzerland	KZ	Kazakhstan	SI	Slovenia
CI	Côte d'Ivoire	ш	Liechtenstein	SK	Slovakia
CM	Cameroon	LK	Sri Lanka	SN	Senegal
CN	China	LR	Liberia	SZ	Swaziland
CS	Czechoslovakia	LT	Lithuania	TD	Chad
CZ	Czech Republic	LU	Luxembourg	TG	Togo
DE	Germany	LV	Latvia	TJ	Tajikistan
DK	Denmark	MC	Monaco	TT	Trinidad and Tobago
EE	Estonia	MD	Republic of Moldova	UA.	Ukraine
ES	Spain	MG	Madagascar	UG	Uganda
FI	Finland	ML	Mali	US	United States of America
FR	France	MIN	Mongolia	UZ	Uzbekistan
GA	Gabon	MR	Manritania	VN	Viet Nam

#### BENCH TOP UV-ACTIVATED ODOR FILTRATION DEVICE

5

10

15

20

25

30

#### Background of the Invention

#### Field of the Invention

The present invention relates to an apparatus and method for removing odors, or other air-borne contaminants, from air.

#### Discussion of the Art

Air in enclosed spaces becomes stale or develops odors owing to contamination by substances such as volatile organic compounds, mold and inorganic compounds. In hospitals, anesthetic gases and toxic sterilizing compounds, such as ethylene oxide, can accumulate in the air. In smaller spaces, such as the interiors of cars, trucks and aircraft, accumulation of contaminants, for example from construction or furnishing material, can be more of a problem.

Tobacco combustion also contributes to contamination.

Treatment of contaminated air has focused on removal of solid particles, such as by filtration, adsorption onto activated carbon or electrostatic precipitation. Such solid removal merely transfers the contaminant from one place to another. In the case of activated carbon, which is a common choice for such removal, the contaminants are adsorbed onto the carbon surface which eventually becomes saturated and must be replaced. Some contaminants, such as low molecular weight gaseous compounds, are not satisfactorily adsorbed or are easily desorbed. Further, efficiency of contaminant removal decreases at elevated temperatures.

Of the many requirements of an air remediating apparatus, a few are of fundamental importance and are not achieved by commercially available systems. These requirements are the ability to handle a variety of contaminants simultaneously at room temperature and under prolonged, long term operation.

2

Recently photocatalysis has been used to remove contaminants. Contaminated air is passed over or through a photocatalyst irradiated with a suitable light source. Anatase titanium dioxide (TiO<sub>2</sub>) is a common form of photocatalyst and is suitably irradiated with UV light at room temperature. Photocatalytic treatment using titanium dioxide removes low molecular weight organic and inorganic gaseous compounds faster than activated carbon at continuous operation and has the advantage that the contaminants are generally broken down into harmless, odorless or less toxic compounds.

5

10

15

20

25

30

U.S. Patent No. 5,045,288 (Raupp et al.) discloses an apparatus which uses photocatalysis to remove contaminants from water. The water is pumped through a bed of photocatalyst irradiated by UV light through UV-transparent walls of a reaction vessel. Although such an arrangement might be modified for use with air instead of water, the large reaction chamber, which is needed for the catalyst bed and the loose catalyst, make such an apparatus unsuitable for small spaces such as rooms. Further, the back-pressure or resistance created by the catalyst impedes the treatment of large volumes of contaminated air and therefore would require powerful equipment to be effective for contaminant removal.

U.S. Patent No. 4,892,712 (Robertson et al.) discloses an apparatus for removal of contaminants from fluids which uses photocatalysis. The apparatus surrounds a UV tube with a cylindrical matrix containing the photocatalyst. The UV tube and matrix are in turn enclosed in a cylindrical fluid-tight jacket having a small fluid inlet tube, perpendicular to the longitudinal axis of the cylindrical jacket, at one end of the apparatus and a corresponding fluid outlet tube at the other end of the apparatus. Although the apparatus is described as being useful for both gases and liquids, this apparatus would not be effective for removing contaminants quickly from the air in a room or laboratory. The back-pressure generated by moving air along the length of the matrix would require a pump or fan inconveniently large.

There is a need for arrangements which are suitable to decontaminate or remove odors from small spaces such as rooms in hospitals, houses or laboratories or interiors of vehicles such as cars, trucks or airplanes. To be suitable for smaller spaces, the decontaminating equipment should be able to treat large volumes of air quickly without being too large or cumbersome. Preferably such equipment could be placed on a bench or table and quickly remove unpleasant or harmful contaminants.

#### **Summary of the Invention**

According to one aspect of the invention there is provided an apparatus for purifying air comprising a housing having an air inlet and an air outlet;

a circulating means, mounted in the housing, to circulate air from outside the housing through the inlet along an air flow path inside the housing and out of the outlet:

a planar filter means disposed across the path of circulating air, said

15 filter means comprising a photocatalyst fixed to a fibrous porous support;

and a light source mounted in the housing to direct light onto the filter means to activate the photocatalyst.

#### **Brief Description of the Drawings**

20

30

The invention will be further described with reference to the accompanying drawings showing, by way of example, embodiments of the invention and elements thereof.

Figure 1 is a partially cut-away schematic side view of an apparatus 25 according to the invention;

Figure 2 is a front view of the embodiment of Figure 1;

Figure 3 is a partially cut-away schematic side view as in Figure 1, but showing the pivoted rear portion in an open position;

Figure 4 is an exploded view of a filter cartridge containing a filter suitable for use in the apparatus of Figure 1;

4

Figures 5 and 6 are front and side views respectively of the filter cartridge of Figure 4:

Figures 7, 8 and 9 are exploded views of different embodiments of a filter suitable for use in the apparatus of Figure 1.

Detailed Description of the Invention

5

10

15

20

25

30

In a preferred embodiment the air flow path has an upstream portion in which air flows in one direction and an adjacent downstream portion in which air flows in the opposite direction and the planar filter means is disposed in the housing such that the filter means extends across the upstream and downstream portions.

The housing provides a path for the air to circulate and a structure on which the other elements such as the light source and the circulating means can be mounted. The housing may be plastic or metal or other suitable structural material. Besides the inlet and outlet, the housing should be reasonably airtight.

The inlet should be big enough to accommodate the filter means. If the inlet is larger than the filter means then air would be allowed to pass without being exposed to the photocatalyst. If it is smaller than the filter means this would waste some of the available area of the filter. The size of the filter means should be big enough to permit suitable air flow. The air inlet may have an additional filter means, upstream of the photocatalyst filter means, such as is used in furnaces, to remove dust and prevent clogging of the photocatalyst.

Since air cannot flow as fast through the filter means as through an unobstructed outlet of the same dimensions, the outlet may be made smaller than the inlet to equalize the system. In an apparatus tested, a 33% size difference was used (0.31 m<sup>2</sup> (48 in.<sup>2</sup>) inlet and 0.23 m<sup>2</sup> (36 in.<sup>2</sup>) outlet).

A protective grille is preferably placed on the outside of the inlet and the outlet. The grille should not substantially interfere with the air flow.

The light source should supply light at sufficient intensity and wavelength band to energize the photocatalyst so that reaction or breakdown of the

5

contaminants is effected. A wide range of light sources can be used. Suitable light sources include: fluorescent blacklights, incandescent blacklights, xenon lamps and low, medium and high pressure mercury lamps. However, if the photocatalyst is titanium dioxide, a source of UV, such as a blacklight having a wavelength band of from 300 to 440 nm, is preferred. Whilst shorter wavelengths such as around 254 nm have been found to be efficient, it is preferred, for safety reasons, to use light in the UV A region. A wavelength band of around 350-380 nm is suitable. A wavelength of 366 nm is preferred.

5

10

15

20

25

30

The intensity of light is governed by the type and number of light sources. For titanium dioxide it has been found that lower power 15 watt mercury bulbs, with a peak output at wavelengths of 313 and 366 nm, are adequate. Such bulbs provide safety and economy. Using such bulbs, it has been found that whilst there is a significant increase in reaction speed (measured by the reduction in contaminants as a function of time) using 2 bulbs rather than 1 bulb, the increase obtained by using 3, rather than 2 bulbs, is less significant. Therefore for domestic applications, 2 bulbs may be preferred for the sake of economy.

The light source is arranged on one side of the filter means. That is, the light source is not surrounded by the filter means. This permits the filter means to have a larger area in the path of the air and the light from the light source radiates to reach the exposed area of the filter means. Preferably a reflecting layer is disposed behind the light source to reflect escaping light back to the filter means.

Any photocatalyst which effectively reduces or removes contaminants may be used. Suitable photocatalysts are TiO<sub>2</sub>, ZnO, CdS, WO<sub>3</sub>, SnO<sub>2</sub>, ZrO<sub>2</sub>, Sb<sub>2</sub>O<sub>4</sub>, CeO<sub>2</sub> and Fe<sub>2</sub>O<sub>3</sub>. A preferred photocatalyst is the anatase form of TiO<sub>2</sub>.

Optimizing odor or contaminant removal will be a balance between conflicting needs such as increasing efficiency on the one hand and increasing air flow and reducing costs on the other hand. For a given photocatalyst, efficiency of contaminant removal will be affected by: the intensity and wavelength of the light; the surface area of the photocatalyst; the volumetric air flow; and the pressure differential across the filter means.

6

The air flow determines the residence time or the time the contaminants are in contact with the photocatalyst. With regard to residence time, the longer the contaminated air is in contact with the photocatalyst, the higher the probability that the contaminants will decompose. On the other hand, it is desirable to process as much air as quickly as possible to reduce the level of contaminants.

The circulation means is conveniently a fan although other means, such as pumps, or convection might be used. It is only required that the means ensures movement of the air over the photocatalyst.

5

10

15

20

25

30

In a preferred embodiment the inlet is in the top of one side the front of the housing and the outlet is at the end of a tunnel protruding out from the bottom of the same side of the housing. This tunnel gives a convenient location for the fan and, by increasing the area of the bottom of the apparatus (the "footprint"), gives stability to the apparatus so that it will not readily tip over. Such a design also permits a further preferred feature in which the filter can extend substantially the whole height of the apparatus. Incoming air is then exposed to the top section of the filter, guided around in a U-turn by the curved configuration of the back of the housing and is then again exposed to photocatalysis on the bottom section of the filter on its way through the tunnel to the outlet. A plenum is used to separate top and bottom sections of the filter in this arrangement.

Other arrangements are, of course, possible. For example, the housing could be constructed so that the circulating air travels in an essentially straight path from an inlet on one side of the housing to an outlet at the other side of the housing. In such an arrangement the housing might conveniently be rectangular in shape. The filter means could be mounted adjacent either the inlet or the outlet or even two filter means could be used one located adjacent each of the inlet and outlet. The circulating means could then be located centrally between the inlet and outlet. Thus it will be seen that various arrangements are possible.

The filter means includes a filter which may be flat or pleated. The filter essentially comprises two components, the support and the photocatalyst. The support is fibrous and should be sufficiently porous to permit adequate air flow.

5

10

. 15

20

25

7

Preferably the support has a large surface area and is transparent to the light used or absorbs little of the light. The photocatalyst should be arranged on the support to maximize the surface area exposed to the circulating air. The photocatalyst could be in the form of a coating on the support prepared conventionally by contacting the support with a coating composition of the photocatalyst in a liquid medium. An effective way to maximize the surface area is to apply the photocatalyst in the form of a finely divided powder to a fibrous non-woven support which is capable of holding the photocatalyst particles electrostatically, such as an electret material. Precharged electret fibers are preferably formed from a film of a dielectric polymer that is capable of being corona charged and then fibrillated. Suitable film forming dielectric polymers include polyolefins, such as polypropylene, linear low density polyethylene, poly-1-butene, polytetrafluoroethylene, polytrifluorochloroethylene; or polyvinylchloride; aromatic polyarenes, such as polystyrene; polycarbonates; polyesters; and copolymers and blends thereof. Preferred are polyolefins free of branched alkyl radicals and copolymers thereof. Particularly preferred are polypropylene and polypropylene copolymers.

Various functional additives known in the art can be blended with the dielectric polymers or copolymers such as poly(4-methyl-1-pentene) as taught in U.S. Pat. No. 4,874,399, a fatty acid metal salt, as disclosed in U.S. Pat. No. 4,789,504, or particulates as per U.S. Pat. No. 4,456,648. Such electret fibers will electrostatically fix particulates. Blown fibers, particularly blown polypropylene microfibers are preferred as the support material. It is further preferred that the fibrous support is polypropylene having a weight range of from 20 to 80 gm/m², more preferably in the range of from 30 to 60 gm/m².

The preferred support material, blown polypropylene microfibers, is available commercially in rolls having a thickness of 0.127 mm (5 thousandths of an inch) in compressed form and 0.508 mm (20 thousandths of an inch) in uncompressed form. For the present invention it is preferred to "fluff" the material giving an uncompressed thickness of 1.524 mm (60 thousandths of an inch). This

WO 96/37281

5

10

15

20

25

material, after having been loaded with the photocatalyst, is preferably used as a double or triple layer.

A filter means in which the photocatalyst is electrostatically fixed to the support, is readily prepared by contacting the finely divided photocatalyst powder with the support and removing any loosely adhered particles, or excess particles, by vacuuming.

A photocatalyst in powder form can also be loaded on the support by coating the support with, for example, a thin water-based acrylic adhesive and then contacting the coated support with the photocatalyst powder. Titanium dioxide can be loaded on a polypropylene support using this technique.

The photocatalyst powder should generally have a particle size of less than 1 mm or the support may not be able to hold the particles electrostatically. Further, a smaller particle size would normally provide a larger surface area and hence increase the reaction speed. Particle sizes ranging from 1 nm to 1 mm (1000 microns) are suitable, preferably from 2 nm to 1000 nm.

The filter should generally provide a surface area of photocatalyst of at least  $1 \text{ m}^2/g$ , preferably at least  $5 \text{ m}^2/g$ . When the photocatalyst is in powder form and is electrostatically fixed to the support, substantially all the surface area of the photocatalyst is available for reaction. Suitable titanium dioxide photocatalysts in powder form are available from Degussa under the trade designation "P 25", which has a surface area of about  $50 \text{ m}^2/g$ , and from Hombikat under the trade designation UV100, which has a surface area of about  $250 \text{ m}^2/g$ . However, it is to be noted that when the particles are extremely fine they may clump together resulting in a decrease of the predicted surface area.

A filter means prepared by coating a composition of a titanium dioxide photocatalyst in a liquid medium onto a support would typically have a surface area of from about 10 to about 15 m²/g.

Using an electrostatically bound powdered photocatalyst on a fibrous support, up to 50% by weight of photocatalyst can be loaded on the support based

on the weight of the support. A preferred loading range is from 10 to 25% by weight.

By use of an activated photocatalyst, such as titanium dioxide, both organic and inorganic compounds can be decomposed. For example: hydrocarbons can be decomposed to carbon dioxide and water; and hydrogen sulfide can be decomposed to elemental sulfur. The reactions which occur do not consume the photocatalyst which may be continually re-activated by the light.

5

10

15

20

25

The filter means may additionally comprise a layer of an adsorbent, such as activated carbon, on at least one other fibrous porous support layers to benefit from the combination of both photocatalytic and adsorptive contaminant reduction or removal. If such an additional layer is used, it should be on the side of the photocatalyst layer opposite the light source to avoid blocking the light and obstructing activation of the photocatalyst.

Since the filter means is disposed across the path of the circulating air, when the apparatus is in operation, a back pressure is generated. The back pressure is a function of the pressure drop across the filter. To give an example, in a domestic furnace, the pressure drop across a filter would be around 3.81 mm (0.15 inches) of water at a face velocity of 1.524 m/s (300 ft/min). In such a domestic furnace, if the pressure drop across the filter rose to 12.7 mm (0.5 inches) of water at 1.524 m/s (300 ft/min), the fan might not have enough power to circulate air.

For table top or bench top applications, such as preferred for an apparatus of the present invention, the pressure-drop across the filter should preferably be in the range of 7.62 mm to 12.7 mm (0.3 to 0.5 inches) of water at a face velocity of 0.381 m/s (75 ft/min), 15.24 mm to 25.4 mm (0.6 to 1.0 inches) at 1.524 m/s (300 ft/min). Ideally, the pressure drop should be as low as possible for a given air flow, consistent with achieving effective contact of the air with the photocatalyst.

Preferably, the air flow will range from a face velocity of from 0.0508 m/s to 3.556 m/s (10 ft/min to 700 ft/min), more preferably from 0.127 m/s to 1.778 m/s (25 ft/min to 350 ft/min).

1.0

15

20

25

30

Using the preferred polypropylene support material a thickness of from 3.048 mm (120 thousandths of an inch), for a double layer, to about 3.302 mm (130 thousandths of an inch) for a compressed triple or multiple layer, is suitable.

It is preferred that the support is pleated or corrugated. This pleated construction permits a greater surface area without significantly increasing the back pressure or impeding the air flow. To assist in constructing such a pleated structure the support can be mounted on a backing material which can be configured to hold the desired shape. The backing material should not interfere with air flow or the energization of the photocatalyst. Expanded metal or scrim is suitable as a backing material. Scrim is a net of material such as polypropylene and is available for example, from the Conwed Corp. A form of scrim commercially available has a thickness of about 0.254 mm (10 thousandths of an inch). The backing layer may be coated with a layer of pressure-sensitive adhesive to hold the support. The filter means may have multiple layers of the support, for example a layer of support on each of a plurality of backings, or a single backing might have a layer of support on each side so the backing layer is actually in between the support layers. Of course, having more layers will impede air flow. A single or double-layer, pleated, is preferred.

The apparatus can be used to remove various odors and contaminants, for example, low molecular weight organic gases such as isobutane, propane, diethylether and acetaldehyde and higher molecular weight gases such as ethylpropionate (fruity rum smell), allylbutyrate (peach, apricot smell) and propylbutyrate (sweaty, rancid smell). An important contaminant which can be removed is hydrogen sulfide. It has been said that 80 percent of malodorous gases contain hydrogen sulfide.

With reference to Figure 1, an apparatus for purifying air according to the invention includes a housing 1 which is generally L-shaped with a curved back. The housing is made of structural plastic material but could be made of any other suitable structural material such as metal. The front of the housing has a top portion comprising an air inlet 2 and bottom portion having an extension generally

11

indicated by 3 terminating in an air outlet 4. A fan 5 is mounted in the extension 3. The extension 3 provides a convenient location for fan 5 and additionally provides a stable design. A filter means in the form of a generally planar filter cartridge 6 is mounted within the housing adjacent inlet 2 and extends from the top of the housing to the bottom of the housing generally coplanar with inlet 2. It can be seen that the filter cartridge 6 extends across both the upstream and downstream air flow passages such that the cross-sectional area of the cartridge is substantially equal to the sum of the areas of the upstream and downstream passages. Disposed in cartridge 6 is a filter 7 which comprises one or more layers of a support having a photocatalyst fixed thereon. A plenum 8 separates the filter cartridge 6 into an upper filter section and a lower filter section so that circulating air, shown by arrows 9, 10, 11 and 12, passes through the upper section of the cartridge 6 after entering inlet 2 and then passes through the lower section on its way to the outlet 4. In this embodiment the air is thus exposed twice to the filter. The preferred configuration of filter 7 shown in Figure 1 is a pleated arrangement in which filter 7 comprises a plurality of planar faces each disposed at an angle to the general direction of the air flow. The filter will be discussed in more detail hereinafter.

5

10

15

20

25

30

UV lamps 13 act as the light source and are mounted in the side walls of housing 1 on one side of filter cartridge 6 and between filter cartridge 6 and the back of the housing. UV lamps are a preferred light source for titanium dioxide as photocatalyst. The lamps shown are conventional cylindrical UV lamps and extend from one side wall of the housing to the other side wall. The positioning of lamps 13 is not critical, provided they can energize the photocatalyst effectively. The lamps are activated by a switch 14 mounted at the top of housing 1 for easy access, and a ballast 15, mounted at the bottom of the housing for stability.

The back of housing 1 comprises a shroud 16 which is pivoted at pivots 17 at the top of the housing and can be opened upwardly for easy access to the inside of the housing. A lock 18 and catch 19 fasten the pivoted shroud 16 in its closed position. A reflective surface 20 of polished aluminum on the inside surface of shroud 16 and the back of housing 1, redirects light escaping from the lamps 13 to

10

15

20

25

30

the filter 7 to maximize efficiency. Polished aluminum is the preferred material for reflection of UV light.

Figure 2 shows a front view of the apparatus of Figure 1. Housing 1 is provided with a protective inlet grille 30, disposed across the inlet 2 (not shown in this Figure), and protective outlet grill 31, disposed across the outlet 4 (not shown in this Figure).

Figure 3 is another side view of the apparatus shown in Figures 1 and 2 and is the same as Figure 1 except that it shows the pivoted shroud 16 in an open position.

In operation, fan 5 draws outside air through inlet 2 and through the upper portion of filter 7. The photocatalyst on filter 7 is energized by the UV lamps 13 which causes breakdown and degradation of contaminants in the air in contact with it. The air continues to be drawn through housing 1 around the back and through the lower section of filter 7 where further breakdown and degradation are effected in the lower section of the filter. Reaction products and unreacted gases and air then pass through extension 3 and are expelled from outlet 4.

Figures 4, 5 and 6 show the filter cartridge 6 of Figure 1 in more detail.

Cartridge 6 has an outside casing of two detachable sections 40 and 41 which contain the filter 7. The casing permits easy handling and protection of filter 7. In the embodiment shown in the drawings, filter 7 has a pleated or corrugated shape.

Plenum 8 divides the upper (inlet) section from the lower (outlet) section of the filter and prevents air from being permitted to bypass the filter. Narrow lips 42 and 43 extend across the front and back faces of the casing sections to comfortably retain the filter in the casing.

Figures 7, 8 and 9 are exploded views of three types of filter suitable for use in the present invention. Figure 7 is a view of the filter 7 shown in Figures 4, 5 and 6. Support 71 is a double layer of blown polypropylene microfibers loaded with a photocatalyst powder of titanium dioxide electrostatically fixed to the fibers. Backing layers 72 and 73 are expanded metal mesh which have been formed into a pleated or corrugated shape. To achieve the filter 7 (shown in

15

20

25

30

Figure 1) the support layer 71 is pressed between expanded metal mesh layers 72 and 73 forcing the pliable fibrous support layer 71 to adopt the same pleated shape as expanded metal mesh layers 72 and 73.

In Figure 8, support 81 is a single layer of the same titanium dioxide loaded polypropylene fibers as shown in Figure 7. Since only a single backing layer 82 is used, support 81 is fixed to the backing layer 82 by applying a coating of pressure-sensitive adhesive to either the backing layer 82 or the support layer 81.

Figure 9 is the same as Figure 7 except that support 91 is a single layer (as in Figure 8) between the two backing layers 92 and 93. A further difference is that an additional support layer 94 is present. The additional support layer may be an additional layer of polypropylene fibers loaded with titanium dioxide photocatalyst or it may be a porous layer containing a conventional adsorbent such as a layer of active carbon fixed to a porous web of polyester fibers.

An apparatus according to the invention was constructed by modifying an LP 1500 Bionaire air purifier. The UV lamps were 15 watt, 0.3048 m (twelve inch) RPR-3000 lamps (such as used in a Rayonet Photochemical Reactor) obtained from the Southern Ultraviolet Co. The fan had two speeds. The high speed or high flow setting provided a flow rate of 1080 L/min. The low speed setting provided a flow rate of 604 L/min. In the apparatus used these settings correspond to face velocities of 0.9144 m/s (180 ft/min) (high flow) and 0.508 m/s (100 ft/min) (low flow).

The modified Bionaire LP 1500 air purifier which was used in the examples, and represented schematically by Figures 1, 2 and 3, had a height of 0.222 m (8.75 inches), a depth of 0.27 m (10.625 inches) and a width of 0.31 m (12.25 inches). These dimensions give a compact device with a modest "footprint" suitable for a bench or desk top.

The filter cartridge used had a width of 0.2921 m (11.5 inches) and a height of 0.197 m (7.75 inches). The plenum was 0.086 m (3.38 inches) from the bottom of the cartridge. The cartridge was 0.0381 m (1.5 inches) thick and the distance from one pleat to the next, along the plane of the pleat was 0.04 m (1.58 inches).

14

Thus a filter with eight pleats had 16 pleat faces making it comparable to a flat unpleated filter spanning  $0.4064 \text{ m} \times 0.04 \text{ m} (16 \times 1.58 \text{ inches})$ . The lip of the cartridge was about 0.0127 m (0.5 inches) all round.

#### 5 EXAMPLES

10

15

20

25

Examples representing the present invention, a comparative example, and a reference (Philips) were tested as follows:

The apparatus was placed in an air-tight chamber 0.609 m by 0.609 m by 0.609 m (2 feet by 2 feet by 2 feet) (227 liters) except if indicated otherwise. A known amount of contaminant was introduced into the chamber. For testing purposes, the initial concentration of contaminant was sometimes adjusted to as much as 1500 to 2000 ppm. (In normal use, concentrations would be about 1-20 ppm). After a few minutes to allow for equilibration, the apparatus was activated and samples removed with a syringe at regular intervals. The samples were analyzed with a Perkin Elmer Autosystem Gas chromatogram - 9000 equipped with a methanizer. The temperature of the flame ionization detector was reduced from 350°C to 225°C when testing for isobutane and propane, to improve accuracy. For hydrogen sulfide, the concentration was measured with a confined space monitor available from 3M under the trade designation "Dynatel CSM 500".

The apparatus was compared to a Philips Clean Air System 75 (as a reference) which is considered to be an industry standard. The Philips device is designed to remove odors and has two activated carbon filters. It has a flow rate of 3136 L/min. Air is drawn in from the sides of the device and exhausted from the top of the device.

The apparatus was tested using the following compounds as contaminants: isobutane, hydrogen sulfide, propane, diethyl ether, acetaldehyde, ethyl propionate, allyl butyrate and propyl butyrate. The contaminant used is indicated in each of Tables 1 to 15B below. The filter tested and flow rate used for the apparatus/filter combinations is indicated in each of Tables 1 to 15B.

The apparatus tested had 3 UV bulbs with a wavelength of 313 nm each, unless otherwise indicated. Notably, the number of bulbs (with the same wavelength) was varied in Example 6 and the wavelength (with the same number of bulbs) was varied in Table 7.

The absence of a contaminant concentration reading in the tables below indicates that such a reading was not taken for the time period designated in the column of the table.

#### Preparation of Filters

10

15

20

25

5

#### Filter #1

A web of blown polypropylene non-woven fibers was needle-tacked into a web. The web was shaken in a bag with excess titanium dioxide powder (available from Degussa under the trade designation "P 25") until the web was saturated with the powder. The titanium dioxide loaded web was vacuumed in a fume hood or with a portable vacuum to remove loose powder. The web was folded and placed on a pleated wire mesh backing layer (0.381 mm (0.015 inches) thick and 31.75 mm (1.25 inches) diamond expanded metal mesh) having eight pleats or corrugations. A second pleated wire mesh backing layer was pressed onto the other side of the web. The filter was placed in a cardboard frame (0.3098 m (12 inches) by 0.2032 m (8 inches)) for ease of handling.

#### Filter #2

The procedure for Filter #1 was followed except that only one layer of polypropylene web was used and the web had a pleated wire mesh backing on only one side. The web was fixed to the backing by a coating of pressure-sensitive adhesive.

16

#### Filter #3

A sheet of glass fiber was used instead of the blown polypropylene fibers. The titanium dioxide was coated on the glass fibers with a sol gel process (such as is used in US Patent No. 4,892,712). Otherwise the procedure was as for Filter #1.

5

#### Filter #4 and Filter #5

The procedure for Filter #1 was followed except that a scrim material was used as backing layer. The scrim was coated with a pressure-sensitive adhesive.

Since the scrim is difficult to pleat, this type of filter was unpleated and flat.

10

15

## Filter #6

The procedure for Filter #1 was followed except that one layer of titanium dioxide loaded web was replaced by a layer of a polyester fiber web coated with an activated carbon adsorbent. This hybrid filter has both a photocatalyst layer and an additional layer of activated carbon on a polyester fiber support.

# EXAMPLE 1

TABLE 1

	Contamin	ant (Isobutane) Concentrat	ion (ppm)		
	Apparatus/Filter Combination				
Time (Hours)	Reference i (Philips)	Example A Filter #1 High Flow (1080 L/min)	Example B Filter #1 Low Flow (604 L/min		
0.00	1473	1361	1406		
0.50	1364	1321	1355		
1.00	1355	1291	1311		
1.50	1347	1259	1264		
2.00	1330	1229	1221		
2.50	1323	1205	1186		
3.00			1172		
3.25	1299				
3,50		1141	1149		
3.75	1291				
4.00		1126	1123		
4.25	1276	1			
4.50		1110			
4.75 .	1265				
5.00		1088	812		
5.25	1258				
5.75	1242				
6.00		1045			
6.75	1208				
6,83			859		
7.00		1004			
7.50		982			
7.75	1192				
22,50			300		
22.75		474			
23.25	923	1			

5

TABLE 2

	Contaminant (Propan	e) Concentration (ppm
	Apparatus/Filt	ter Combination
Time (Hours)	Reference 2 (Philips)	Example C Filter #1 High Flow (1080 L/min)
0.00	1414	1703
0.33		1649
0.50	1362	
0.67		1635
1.00	1348	1619
1.50	1345	1579
2.00	1338	1555
2.50	1327	1531
3.00	1316	1509
3.50	1309	
3.67		1482
4.00	1300	
4.50	1291	
4.75		1408
5.00	1269	
6.00		1261
6.50		1264
20.50	1008	
21.50		815

# EXAMPLE 3

TABLE 3

	Contamin	ant (Diethylether) Concent	ration (ppm)	
	Apparatus/Filter Combination			
Time (Hours)	Reference 3 (Philips)	Example D Filter #1 High Flow (1080 L/min)	Example E Filter #1 Low Flow (604 L/min)	
0.00	1909	1665	1524	
0.50	1870	1392	1306	
1,00	1880	1147	1106	
1.50	1867	986	952	
2.00	1858	864	810	
2,50	1826	741	719	
3,00	1814	654	636	
3,50	1763	568	552	

5

TABLE 4

	Contaminant (Acetalde	hyde) Concentration (ppm)	
F	Apparatus/Filter Combination		
Time (Hours)	Reference 4 (Philips)	Example F Filter #1 High Flow (1080 L/min)	
0.00	2605	2874	
1.00		2065	
2.08	2494	1655	
3.00		1400	
3.33	2386		
4.08		1279	
4.83	2385		

TABLE 5

	Con	ntaminant (Isobutane)	Concentration (ppn	n)
Ī		Apparatus/Filter	Combinations	·
Time (Hours)	Reference 5 (Philips)	Example G Filter #3 Low Flow (604 L/min)	Example H Filter #1 Low Flow (604 L/min)	Example I Filter #2 Low Flow (604 L/min
0.00	1473	1380	1406	1384
0.50	1364	1352	1355	1356
1:00	1355	1314	1311	1335
1.50	1347	1301	1264	1316
2.00	1330	1262	1221	1295
2.50	1323	1265	1186	1256
3.00			1173	
3.17		1232		
3.25	1299			
3.50		1234	1149	1202
3.75	1291			
4.00		1224	1123	
4.25	1276			
4.50		1208		1150
4.75	1265			
5.25	1258			
5.50		1177		1113
5.75	1242			
6.00			912	
6.50		1141		1083
6.75	1208	<u> </u>		
6.83			860	
7.00		1126		1076
7.75	1192			
22.50			301	l
23,00		618		572
23.25	923		4.000	

TABLE 6

	Contaminant (Isobutane) Concentration (ppm)				
		Apparatus/Filter Combinat	ion		
Time (Hours)	Example J	Example K	Example L 1 UV Bulb		
	3UV Bulbs	2 UV Bulbs			
1	Filter #1	Filter #1	Filter #1		
	Low Flow	Low Flow	Low Flow		
	(604 L/min)	(604 L/min)	(604 L/min)		
0.00	1448	1303	1262		
0.50	1370	1215	1232		
1.00	1371		1218		
1.50			1201		
2.00	1257	1198			
3.00		1110	1221		
3.50	1235				
4.00	1215	1070			
4.08			1144		
5.00	1158	1033			
5.17			1122		
5.83			1093		
6.00	1117				
22.00	573	576	770		

# **EXAMPLE 7**

TABLE 7

	Contaminant (Isobuta	ne) Concentration (pp
0.00 0.50 1.00 1.50 2.00 2.50 3.00 3.50 4.00 4.50 5.00 5.50	Apparatus/Fi	Iter Combination
Time (Hours)	Example M 313 nm Filter #1 Low Flow (604 L/min)	Example N 366 nm Filter #1 Low Flow (604 L/min)
0.00	1360	1292
0.50	1311	1263
1.00	1268	1229
1.50	1223	1205
2,00	1181	1169
2.50	1147	1137
	1134	1092
3.50	1112	1083
4.00	1067	1070
4,50		1039
5.00		1029
5.50		990
6.00	882	982
6.50		954
6.83	832	
7.00		936
22.50	291	
23,00		414

5

# EXAMPLE 8

## TABLE 8

10

	Contaminant (Ethyl Propi	onate) Concentration (ppm)	
F	Apparatus/Filter Combination		
Time (Min.)	Reference 6 (Philips)	Example O Filter #1 Low Flow (604 L/min)	
0.0	303	269	
0.5	45	64	
1.0	39	18	
1.5	22	03	
2.0	22	01	

# EXAMPLE 9

TABLE 9

5

	Contaminant (Allyl But	yrate) Concentration (ppm)		
	Apparatus/Filter Combination			
Time (Min.)	Reference 7 (Philips)	Example P Filter #1 Low Flow (604 L/min)		
0,0	84	91		
20	11	22		
40	07	08		
60	05	03		
90	03	01		

## EXAMPLE 10

TABLE 10

10

	Contaminant (Propyl But	yrate) Concentration (ppm)	
	Apparatus/Filter Combination		
Time (Min.)	Reference 8 (Philips)	Example Q Filter #1 Low Flow (604 L/min)	
0	215	154	
20	16	29	
40	10	11	
60	8	5	
90	4	2	

TABLE 11

	Contaminant (Isobuta	ne) Concentration (pp	
	Apparatus/Filter Combination		
	Example R	Example S	
Time (Hours)	Filter #4	Filter #5	
	Low Flow	High Flow	
	(604 L/min)	(1080 L/min)	
0,00	1448	1473	
0.25		1450	
0.50	1402	1431	
0,83		1404	
1,00	1362		
1.33		1356	
1,50	1327		
1,83		1330	
2.00	1289		
2.50	1256		
2.67		1294	
3.00		1247	
3.50	1186	1216	
4.00		1180	
4.50		1158	
4.67	1124		
5.00		1120	
5.50	1077	J	
6.00		1067	
6.50	1030		
7.00	L	1009	
7.17	1003		
22.83		476	
23.00	502		

TABLE 12

	Contaminant (Hydrogen Sulf	de) Concentration (ppr
	Apparatus/Filter	Combination
Time (Hours)	Reference 9 (Philips)	Example T Filter #1 High Flow (1080 L/min)
0.00	40.60	40.60
0.25	39.10	33.30
0,50	37.70	22.60
0.75	.36.20	14.40
1.00	34.80	8.10
1.25	33.30	4,10
1.50	31.90	1.90
1.75	30.40	
2.00	30.30	•

#### **EXAMPLE 13**

TABLE 13

	Contaminan	t (Hydrogen Sulfide) Con	centration (ppm)
ľ		Apparatus/Filter Combina	ation
Time (Hours)	Example U Filter #1 Low Flow (604 L/min) UV On	Example V Filter #1 High Flow (1080 L/min) UV On	Comp. Example A Filter #1 High Flow (1080 L/min) UV Off
0.00	40	41	42
0.25	29	33	42
0.50	25	23	42
0.75	20	14	41
1.00	15	08	41
1.25	12	04	39
1.50	08	02	39
1.75	05		39
2,00			38

5

NOTE: In Examples 12 and 13 the testing chamber was a larger chamber of 510 liters capacity. Also, the hydrogen sulfide concentration was determined with a confined space monitor available from Minnesota Mining and Manufacturing Company under the trade designation "Dynatel CSM-500" which has an electro-chemical sensor for hydrogen sulfide.

10

# **EXAMPLE 14**

Table 14A

	Contaminant (Hydrogen Sulfide) Concentration (ppm)
	Apparatus/Filter Combination
Time (Hours)	Example W
	Filter #6
	High Flow (1080 L/min)
0,00	39.1
0.25	21.2
0.50	9.6
0.75	3.2
1.00	1.0
1.25	0.6
1.50	0.4

5

Table 14B

	Contaminant (Isobutane) Concentration (ppm)
F	Apparatus/Filter Combination
Time (Hours)	Example X
	Filter #6
	High Flow (1080 L/min)
0.00	1484
0.33	1090
0.67	1086
1.00	1080
1.33	1066
1,67	1052
2.00	1039
3.00	968
4.00	947
5.00	906
6.00	884
7.00	857
22.25	642

#### **EXAMPLE 15**

Table 15A

	Contaminant (Hydrogen Sulfide) Concentration (ppm)
H	Apparatus/Filter Combination
Time (Hours)	Example Y
	Filter #6
	High Flow (1080 L/min)
0.00	40.6
0.25	33.3
0,50	26.0
0.75	20.7
1.00	16,2
1.25	12.2
1.50	9.0
1.75	6.4
2,00	4.5
2.25	3.2
2.50	2,6
2.75	2.2
3.00	2.0

Table 15B

	Contaminant (Isobutane) Concentration (ppm)
F	Apparatus/Filter Combination
Time (Hours)	Example Z
` '	Filter #6
	High Flow (1080 L/min)
0.00	1455
0.25	1445
0.75	1431
1.75	. 1366
2.75	1314
3.75	1263
4.75	. 1225
5.75	1189
21,25	748

10

The clean air delivery rate (CADR) is a convenient parameter to compare contaminant removal rates. If  $\ln{(C_A/C_A)}$  is plotted against time, where  $C_{At}$  is the contaminant concentration at a given time and  $C_A$  is the initial concentration, then

the slope of the graph will be the decay constant k. The natural decay constant  $k_n$  is the slope of the same graph for results obtained without photocatalysis or other active removal. The CADR is then calculated as:

$$CADR = (k-k_n)V$$

5 where V is the volume of the test chamber.

Using the above examples, Tables A to E can be generated. The table from which the decay constant k was calculated is indicated below as the "Resource Table" and the example or reference is indicated as "Example Designation".

10

TABLE A

Decay constants (k) and clean air delivery rates (CADR) for different filters tested with isobutane (initial concentration approximately 1400 parts per million).

Resource Table	Example Designation	Filter	Flow Rate (L/min)	k (1/hr)	CADR (L/hr)
5	Ex. G	Filter #3	604	0,03404	6.71
ı	Ex. B	Filter #1	604	0.06894	14.64
1	Ex. A	Filter #1	1080	0.04572	9.37
5	Ex. I	Filter #2	604	0.0382	7.68
11	Ex. R	Filter #4	604	0.04565	9.35
11	Ex. S	Filter #5	1080	0.04948	10.22
14B	Ex. X	Filter #6	1080	0.028096	5.36
15B ·	Ex. Z	Filter #6	1080	0.031244	6.078
1	Reference 1	Reference (Philips)	3115	0.01812	3.10

TABLE B

Decay constants (k) and clean air delivery rates (CADR) for Filter #1 tested with initial concentrations of isobutane at approximately 1400 ppm.

Resource Table	Example Designation	Number of UV Bulbs	Wavelength of UV Bulbs (nm)	k (I/hr)	CADR (L/hr)
6	Ex. K	2	313	0.03585	7.12
6	Ex. L	1	313	0.02225	5.051
. 7	Ex. M	3	313	0.06894	15.65
7	Ex. N	3	366	0.04916	11.16

5

Note: The decay constant (k<sub>n</sub>) for isobutane was determined to be 0.00446 L/hr.

TABLE C

Decay constants (k) and clean air delivery rates (CADR) for low molecular weight gases (initial concentrations of approximately 1500 ppm).

Resource Table	Example Designation	Gas Tested	Filter	Flow Rate (L/min)	k (1/hr)	CADR (L/hr)
2	Ex. C	Propane	Filter #1	1080	0.0343	7.788
2	Reference 2	Propane	Reference (Philips)	3115	0.0155	3,515
3	Ex. E	Diethylether	Filter #1	604	0.2895	65.72
3	Ex. D	Diethylether	Filter #1	1080	0,3036	68.91
3	Reference 3	Diethylether	Reference (Philips)	3115	0.01905	4.324
1 4	Ex. F	Acetaldehyde	Filter#1	1080	0.1974	44.8
<u> </u>	Reference 4	Acetaldehyde	Reference (Philips)	3115	0.01967	4.466

Note: The natural decay constant  $(k_n)$  was not obtained for these gases and was taken to be zero. As a result, the actual clean air delivery rates would be

15 slightly lower than the reported values.

TABLE D

Decay constants (k) and clean air delivery rates (CADR) for higher molecular weight gases (initial concentrations of approximately 100-500 ppm).

5						
Resource Table	Example Designation	Gas Tested	Filter	Flow Rate (L/min)	k (l/min)	CADR (L/min)
8	Ex. O	Ethyl Propionate (fruity rum smell)	Filter #1	604	0.051817	11.76
8	Reference 6	Ethyl Propionate (fruity rum smell)	Reference (Philips)	3115	0.019917	4.52
9	Ex. P	Allyl Butyrate (apricot smell)	Filter #1	604	0.04633	10.52
9	Reference 7	Allyl Butyrate (apricot smell)	Reference (Philips)	3115	0.032433	7.362
10	Ex. Q	Propyl Butyrate (sweaty smell)	Filter #1	604	0.03715	11.08
10	Reference 8	Propyl Butyrate (sweaty smell)	Reference (Philips)	3115	0.0529	12.01

Note: The natural decay constant  $(k_0)$  was not obtained for these gases and was taken to be zero. As a result, the actual clean air delivery rates would be slightly lower than the reported values.

10

TABLE E

Decay constants (k) and clean air delivery rates (CADR) for hydrogen sulfide (initial concentrations of approximately 40 ppm).

Resource Table	Example Designation	Filter	Flow Rate (L/min)	k (1/min)	CADR (L/min)
13	Ex, U	Filter #1	604	0.01868	9.242
13	Ex. V	Filter #1	1080	0.03428	17.2
13	Comp. Ex. A	Filter #1 (UV Off)		0.000919	0.1857
14A	Ex. W	Filter #6	1080	3.305522	28.10
15A	Ex. Y	Filter #6	1080	1.09159	9.28_
12	Reference 9	Reference (Philips)	3115	0.002603	1,045

15

Note: The natural decay constant  $(k_n)$  for hydrogen sulfide was determined to be 0.000555 L/min.

15

#### TABLE F

Filter #4 was tested using hydrogen sulfide as the contaminant with intial concentrations of 40 ppm at both low and high flow. A resource table is not shown for this test, but the calculations for decay constants (k) and clean air delivery rates (CADR) for hydrogen sulfide (initial concentrations of approximately 40 ppm) are shown below.

Filter	Flow Rate (L/min)	k (l/min)	CADR (L/min)
Filter #4	604	0.0202	10.02
Filter #4	1080	0.03118	15.62

It should be noted that for carbon filters the CADR drops (less efficiency) when the flow rate is decreased. For a photocatalytic device of the invention, decreasing the flow rate generally increases the CADR.

The results demonstrate that the apparatus of the invention outperforms the active carbon filter of the Philips device, especially for the removal of low molecular weight gases such as isobutane, propane, acetaldehyde and hydrogen sulfide.

#### WHAT IS CLAIMED IS:

- An apparatus for purifying air comprising a housing having an air inlet and an air outlet:
- a circulating means, mounted in the housing, to circulate air from outside the housing through the inlet along an air flow path inside the housing and out of the outlet;
- a planar filter means disposed across the path of circulating air, said filter means comprising a photocatalyst fixed to a fibrous porous support;
- and a light source mounted in the housing to direct light onto the filter means to activate the photocatalyst.
- An apparatus according to claim I wherein the air flow path has an upstream portion in which air flows in one direction and an adjacent downstream portion in which air flows in the opposite direction and the planar filter means is disposed in the housing such that the filter means extends across the upstream and downstream portions.
- 3. An apparatus according to claim 1 wherein said air-flow path is 20 straight.
  - 4. An apparatus according to claim 2 in which the sum of the cross-sectional areas of the upstream and downstream portions of the air flow path is substantially equal to the cross-sectional area of the filter means.
- 25
- An apparatus according to claim 1, 2, 3 or 4 wherein the photocatalyst is fixed electrostatically to the support.

· WO 96/37281

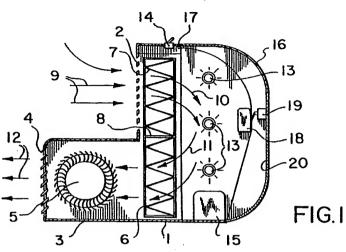
34

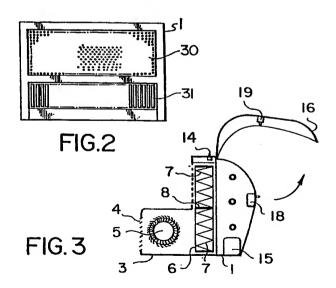
PCT/US96/06305

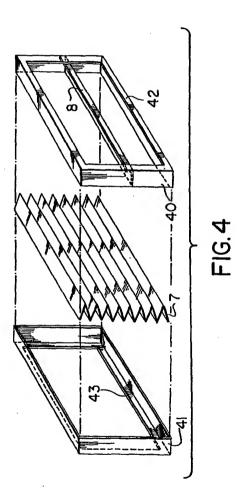
6. An apparatus according to claim 1, 2, 3 or 4 wherein the photocatalyst is powdered titanium dioxide, the source of light is a source of UV light having a peak intensity in the UV B range and the support is blown polypropylene electret fibers.

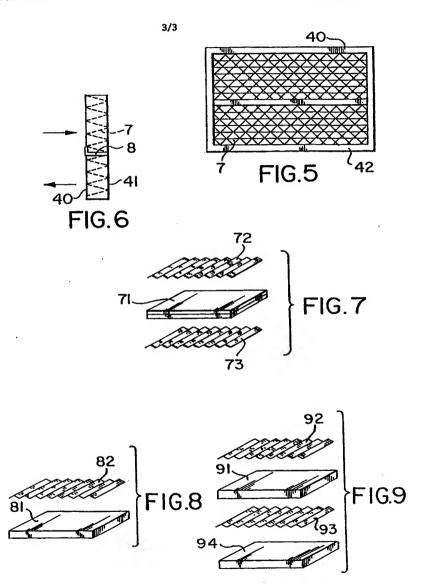
5

- 7. An apparatus according to claim 1, 2, 3 or 4 wherein the support has a structural porous backing of expanded metal mesh and the support has a pleated configuration.
- 10 8. An apparatus according to claim 1, 2, 3 or 4 wherein the filter means further comprises a second fibrous porous support comprising an adsorbent.
  - An apparatus according to claim 8 wherein the adsorbent is activated carbon.









## INTERNATIONAL SEARCH REPORT

			Intonal Appi	ication No
		•	PCT/US 96	/06305
A. CLASSI IPC 6	IFICATION OF SUBJECT MATTER B01D53/86 A61L9/20	-		
	to International Patent Classification (IPC) or to both national cl	assification and IPC		
	SEARCHED ocumentation searched (classification system followed by classification system followed by classifi	ication tymbols)		
IPC 6	B01D A61L			
Documentat	ion searched other than minimum documentation to the extent t	hat such documents are inc	chuded in the fields s	carched
Electronic d	ata base consulted during the international search (name of data .	base and, where practical	, search terms used)	
C DOCUM	IENTS CONSIDERED TO BE RELEVANT			
Category *	Citation of document, with indication, where appropriate, of the	ne relevant passages		Relevant to claim No.
х	EP,A,0 325 133 (HITACHI LTD) 26 see page 5, line 19 - page 5, l see page 7, line 37 - page 7, l claims 1,8,12; figures 8-11	5 July 1989 line 34 line 57;		1-4,8,9
Υ	US,A,5 078 971 (MATUDA MICHIYA January 1992 see column 2, line 66 - column claims 1,2; figures 1,2			1,5-7
Y	US,A,4 892 712 (ROBERTSON MICH/ AL) 9 January 1990 cited in the application see column 4, line 61 - column claim 1; figure 2			1
	<del></del>	-/		
X Furt	ber documents are listed in the continuation of box C.	X Patent family	members are listed	in annex.
"A" docum consid "E" earlier filing; "L" docum which citatio "O" docum other "P" docum later ti	ent which may throw doubts on priority claim(s) or is cited to establish the publication date of another en or other special reason (as specified) ent referring to an oral disclosure, use, exhibition or means ent published prior to the infernational filing date but than the priority date claimed	involve an invent  "Y" document of panel cannot be consid document is com ments, such com in the art.  "&" document membe	icular relevance; the cred novel or canno tive step when the de ticular relevance; the cred to involve an is bined with one or n bination being obvicer of the same paten	claimed invention t be considered to content it staken alone claimed invention the content in the content in the con- tent docu- us to a person skilled to family
	actual completion of the international search  2 August 1996	1	of the international s	earch report
Name and	mailing address of the ISA  European Patent Office, P.B. 5818 Patentlaan 2  NL - 2220 HV Rijavik  Tel. (+31-70) 340-2049, Tx. 31 651 cpo nl,  Fax: (+31-70) 340-3016	Authorized office	-	

Form PCT/ISA/210 (second sheet) (July 1992)

. 1

# INTERNATIONAL SEARCH REPORT Int. Const Application No

Int. ional Application No PCT/US 96/06305

		PC1/05 90	7 00303
	ation) DOCUMENTS CONSIDERED TO BE RELEVANT		Relevant to claim No.
Category *	Citation of document, with indication, where appropriate, of the relevant passages		Kerceant in Gain lace
Y	PATENT ABSTRACTS OF JAPAN vol. 95, no. 001 & JP,A.07 024256 (NIPPONDENSO CO LTD;OTHERS: 01), 27 January 1995, see abstract		1
Y	PATENT ABSTRACTS OF JAPAN vol. 015, no. 038 (C-0800), 30 January 1991 & JP,A,02 273513 (NIPPON ZEON CO LTD), 8 November 1990, see abstract		5,6
Y	PATENT ABSTRACTS OF JAPAN vol. 015, no. 275 (C-0849), 12 July 1991 & JP,A,03 094814 (MATSUSHITA ELECTRIC IND CO LTD), 19 April 1991, see abstract		1,7
	DE,A,42 40 558 (OESTE FRANZ DIETRICH; AALBERS ROBERT (NL); DIETRICH OLGA (DE)) 9 June 1994 see figures 1-7		

1

# INTERNATIONAL SEARCH REPORT | Int. | ional Application No

Information on patent family members

PCT/US 96/06305

			i	
Patent document cited in search report	Publication date	Patent memb		Publication date
EP-A-0325133	26-07-89	JP-A- JP-A- EP-A- US-A- US-A-	1189321 1189322 0476724 4954465 4955208	28-07-89 28-07-89 25-03-92 04-09-90 11-09-90
US-A-5078971	07-01-92	JP~A-	2164420	25-06-90
US-A-4892712	09-01-90	CA-A- EP-A- JP-A- US-A- US-A-	1334519 0306301 1090035 4966759 5032241	21-02-95 08-03-89 05-04-89 30-10-90 16-07-91
DE-A-4240558	09-06-94	NONE		